Femtosecond pulses from the Advanced Light Source: A new tool for ultrafast time-resolved x-ray spectroscopy

H.H.W. Chong[1,3], R.W. Schoenlein[1], T.E. Glover[4], P.A. Heimann[4]
C.V. Shank[1], A. Zholents[2], M. Zolotorev[2]

[1]Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA
[2]Accelerator and Fusion Research Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA
[3]Applied Science and Technology Graduate Group, University of California Berkeley, California 94720, USA
[4]Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA

INTRODUCTION

An important new scientific frontier is the application of x-ray pulses to investigate ultrafast structural dynamics (i.e. atomic motion and rearrangement) associated with phase transitions in solids, chemical reactions, and rapid biological processes. The fundamental time scale for such motion is an atomic vibrational period (~100 fs).

Using a novel electron slicing technique[1], femtosecond synchrotron pulses have been generated directly from the electron storage ring at the Advanced Light Source (ALS). We present our most recent measurements of the ultrashort pulses of synchrotron radiation generated at the ALS and discuss prospects for future work at the ALS with this new capability. This synchrotron-based femtosecond x-ray source makes accessible the application of x-ray techniques on an ultrafast time scale to investigate structural dynamics in condensed matter at the ALS.

EXPERIMENTAL METHODS

The synchrotron radiation pulse duration at the ALS is determined by the electron bunch duration of the electrons stored in the storage ring. The electron slicing technique creates ultrashort slices of electrons (~100fs in duration) from a stored electron bunch (~30ps in duration) and uses these ultrashort electron slices in radiating structures to generate ultrashort pulses of synchrotron radiation.

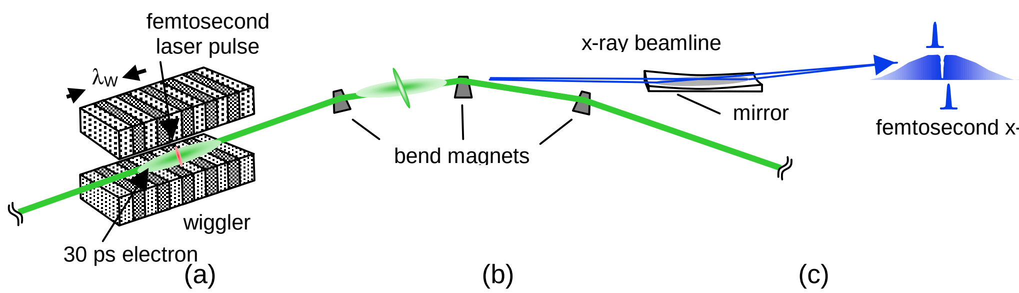

Figure 1. Schematic illustration of experimental apparatus for femtosecond synchrotron radiation pulse generation. (a) An energy modulation is placed on an electron bunch as it co-propagates with an optical laser pulse through a wiggler. (b) In a dispersive section of the ring, the energy modulated electrons are spatially separated into the spatial wings of the main electron bunch. (c) Ultrashort pulses of x-rays generated from the ultrashort slices of electrons can then be extracted at an experimental end-station.

A schematic illustration of the experimental apparatus used to generate femtosecond pulses of synchrotron radiation is shown in Figure 1. An electron bunch and an ultrashort laser pulse co-propagate in a wiggler. The wiggler mediates an energy exchange between the laser pulse and those electrons in the bunch which overlap with the laser pulse, producing an energy modulation on the electron bunch. The energy modulated electrons are separated into the tranverse wings of the main electron bunch in a dispersive section of the storage ring. The energy modulation is translated into a spatial separation as electrons of different energy take different paths due to dispersion. As the electrons travel through a radiating structure, the ultrashort pulses of synchrotron radiation generated by the ultrashort slices of electrons can be selected for use by inserting an aperature at an image plane of the synchrotron light beam.

The optimal conditions for electron slicing in the wiggler require a matching of the spectral and spatial character of the laser light and spontaneous wiggler emission of the electrons. The first of these requirements is best stated as a resonance condition: $\lambda_S=\lambda_W(1+K^2/2)/2\gamma^2=\lambda_L$; the central wavelength of the laser, λ_L , must match the central wavelength of the spontaneous wiggler emission, λ_S; where λ_W is the wiggler period, K is the wiggler deflection parameter, and γ is the Lorentz factor. The spectral bandwidth of the laser light must also match the spectral bandwidth of the wiggler emission averaged over its transverse mode. Matching of spatial character demands the mode of the laser beam match the transverse mode of the spontaneous wiggler emission. Under optimal conditions, the energy modulation produced on the electron bunch is[1]

$$\Delta E = 2\left(A_L A_W \frac{M_W}{\sqrt{2} M_L} \eta_{emit} \right)^{1/2} \cos\phi \quad ,$$

where A_L is the laser pulse energy; $A_W \cong 4.1\alpha\hbar\omega_L K^2/(2+K^2)$ is the energy spontaneously radiated by a single electron passing through the wiggler, α is the fine structure constant, $\hbar$ is Planck's constant, and $\omega_L=2\pi c/\lambda_L$; M_W is the number of wiggler periods; M_L is the laser pulse length in optical cycles (measured full-width at half-maximum, FWHM). The non-zero electron beam size is accounted for by the coefficient $\eta_{emit}\approx 0.7$ in our case.

Experiments were conducted using a mode-locked Ti:sapphire laser at beamline 6.3.2. Femtosecond laser pulses (τ_L=100 fs, A_L=400 μJ, λ_L=800 nm, f_L=1 kHz) were directed into the protein crystallography wiggler (W16) to co-propagate with an electron bunch. The storage ring was operated at 1.5GeV with a corresponding electron energy spread of σ_E=1.2 MeV. The wiggler, with M_W=19 periods and λ_W=16 cm, was adjusted to provide a deflection parameter of $K\cong 13$. The short pulses of synchrotron light generated from electron slices were extracted from beamline 6.3.2 and measured with cross-correlation techniques using visible synchrotron light (~2eV photon energy) and pulses derived from the slicing laser (~3.5eV photon energy). A knife-edge placed in an intermediate image plane of synchrotron beam allowed for the selection of synchrotron radiation generated from the main electron bunch or an electron slice.

RESULTS

A series of cross-correlation measurements were made at various knife-edge positions. Figure 2 presents a progression of knife-edge positions (normalized to the rms horizontal beam size σ_x) starting from the central core of the main electron bunch (collection aperture $\pm 3\sigma_x$) moving outward into the spatial wings. Evident in Figure 2a is the femtosecond "hole" in the main bunch due to an absence of electrons, which were displaced into the spatial wings by electron slicing. Figure 2b displays a measurement of radiation from the spatial wings (knife edge at the $3\sigma_x$) of the main bunch and displays a femtosecond light pulse generated from electrons sliced from the main electron bunch. Figure 2c is even further out in the spatial wings (knife edge at the $4\sigma_x$).

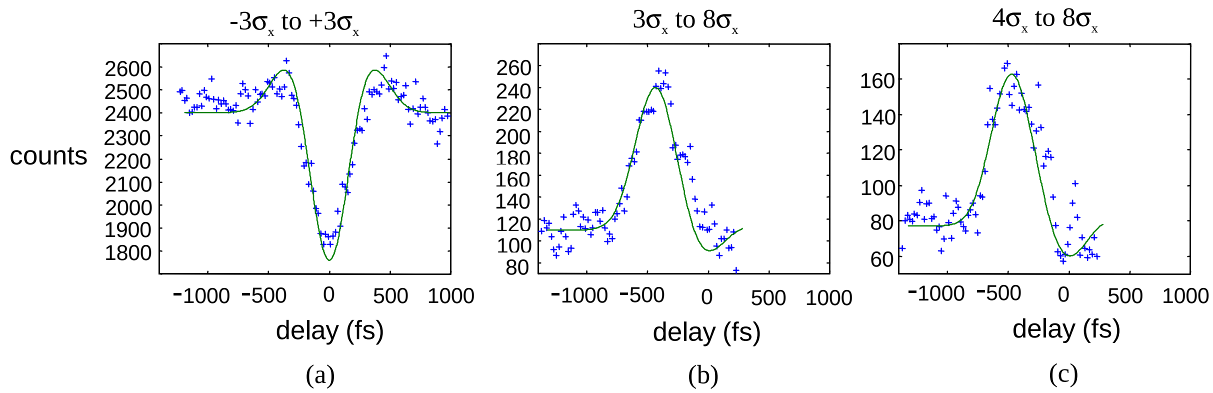

Figure 2. Cross-correlation measurements of visible synchrotron light and femtosecond laser pulse. Vertical axis is photon counts and horizontal axis is time delay relative to slicing laser pulse.

The fits to the data are generated from model calculations of the electron dynamics including energy modulation and dispersion. The model also accounts for diffraction effects due to beamline optics. The calculated electron density distribution is pictured in Figure 3. The model accurately predicts the amplitudes and time delays of the dark and light pulses and gives us confidence that accurate operating specifications can be calculated for different experimental conditions and parameters.

Currently, a dedicated beamline (5.3.1) is being constructed for ultrafast structural dynamical studies using ultrashort (~100fs) x-ray pulses from the ALS. Using a bend-magnet, a femtosecond x-ray flux of ~10^5photons/sec/0.1%BW (for a collection angle of 1 mrad) and an average brightness of ~5x10^7photons/sec/mrad^2/mm^2/0.1%BW at a photon energy of 1 keV is expected. In the future, femtosecond pulses can be generated from a small-gap undulator providing a flux of ~10^7photons/sec/ 0.1% BW and an average brightness of ~10^{11}photons/sec/mrad^2/mm^2/0.1% BW at a photon energy of 1 keV.

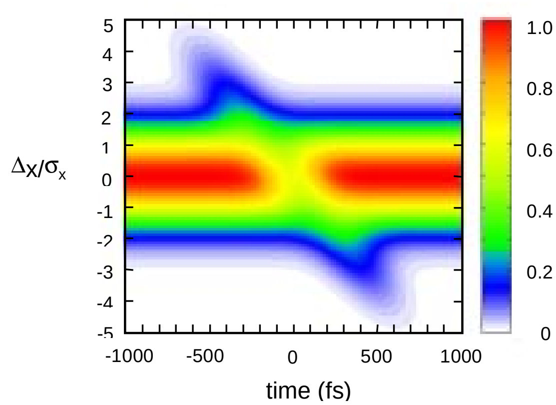

Figure 3. The electron density distrubution at the radiating bend magnet calculated for an electron bunch after interaction with a femtosecond laser pulse in a wiggler and propagating through 1.5 arc sectors of storage ring.

With such x-ray flux and brightness levels available from a femtosecond synchrotron source, a wide range of x-ray techniques including diffraction and EXAFS can to be applied on a 100-fs time scale to investigate atomic motion associated with phase transitions in solids, making and breaking of bonds during chemical reactions, and possibly ultrafast biological processes.

REFERENCES

1. A.A. Zholents, M.S. Zoloterev, *Phys. Rev. Lett.* **76**, 912-915 (1996).

This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Science Division, of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

Principal investigator: Robert W. Schoenlein, Material Science Division, Ernest Orlando Lawrence Berkeley National Laboratory. Email: rwschoenlein@lbl.gov. Telephone: 510-486-6557.